

## Supporting Information

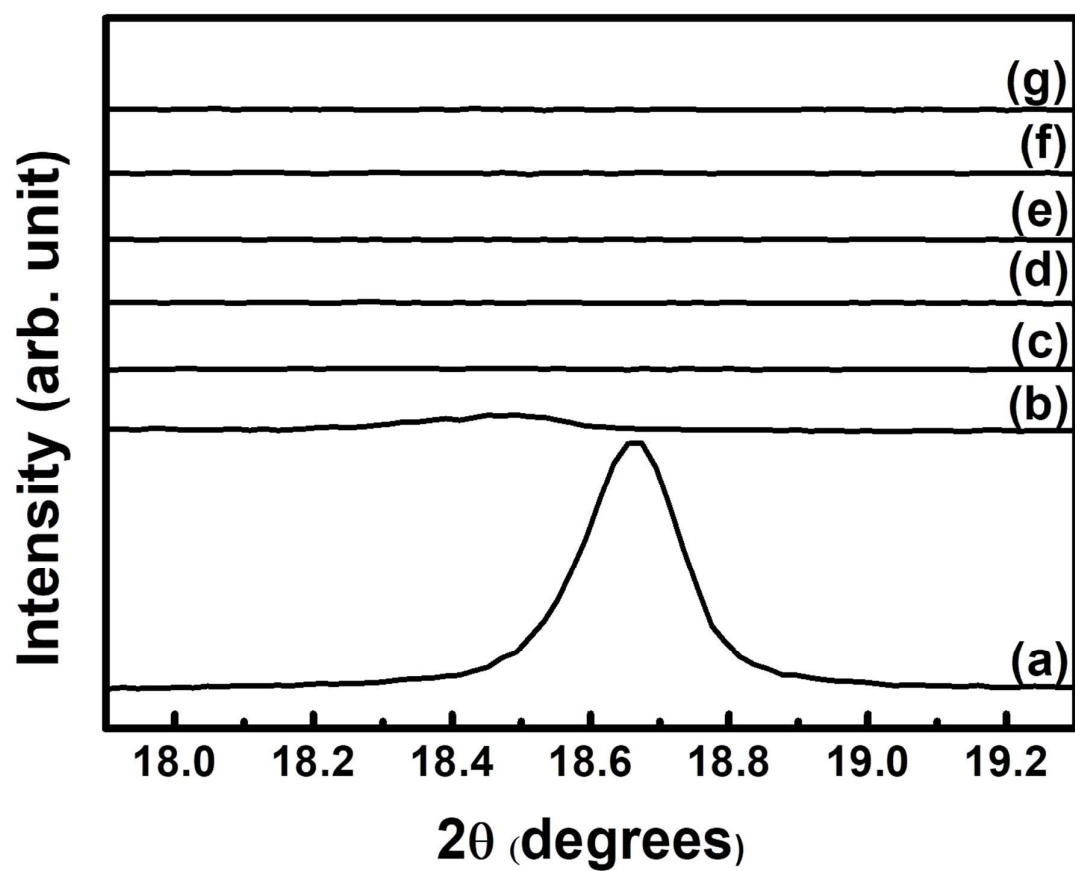
### **Phase transformation from brucite to highly crystalline layered double hydroxide through a combined dissolution-precipitation and substitution mechanism**

Jinseop Shin<sup>1</sup>, Chan-Ju Choi<sup>1</sup>, Tae-Hyun Kim<sup>1,2</sup>, Jae-Min Oh<sup>1\*</sup>

Affiliations

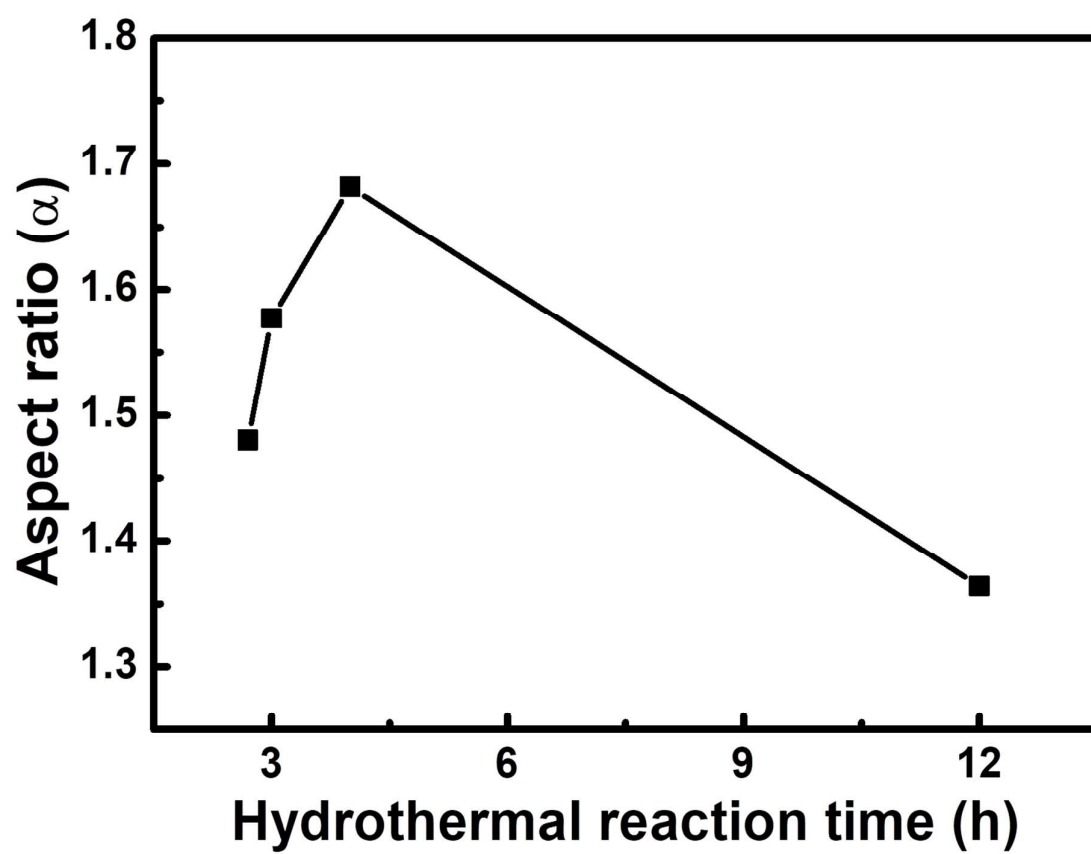
<sup>1</sup> Department of Chemistry and Medical Chemistry, College of Science and Technology, Yonsei University, Wonju 26493, Republic of Korea

<sup>2</sup> Department of Physics, Chemistry and Pharmacy, University of Southern Denmark, Campusvej 55, 5230 Odense M, Denmark

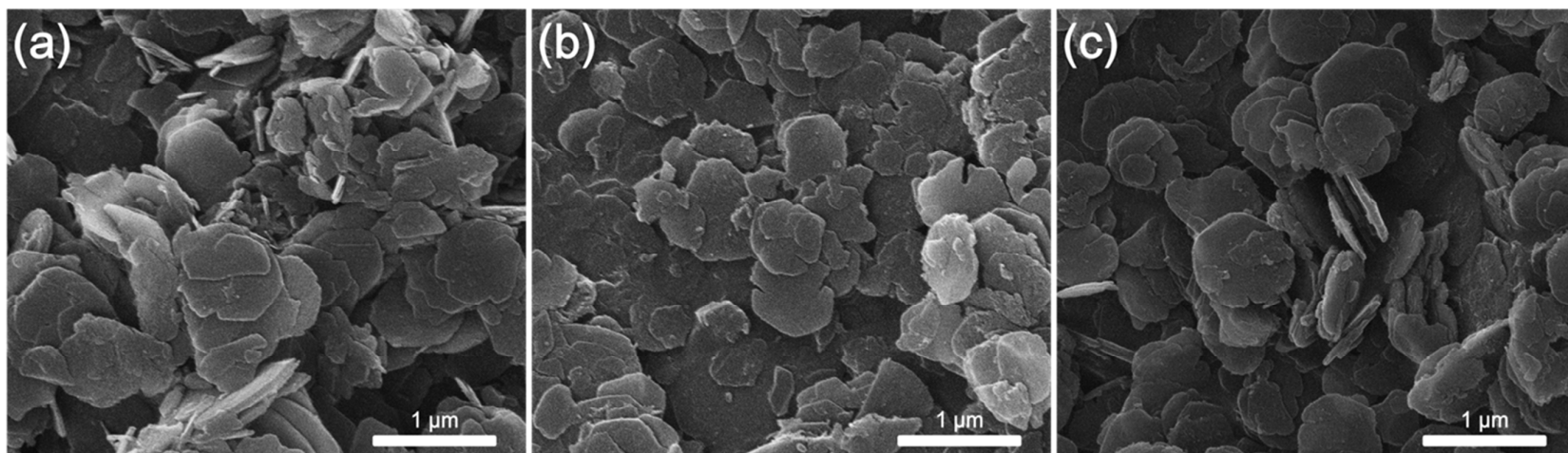


**Figure S1.** Magnified X-ray diffraction peaks for (001) of brucite

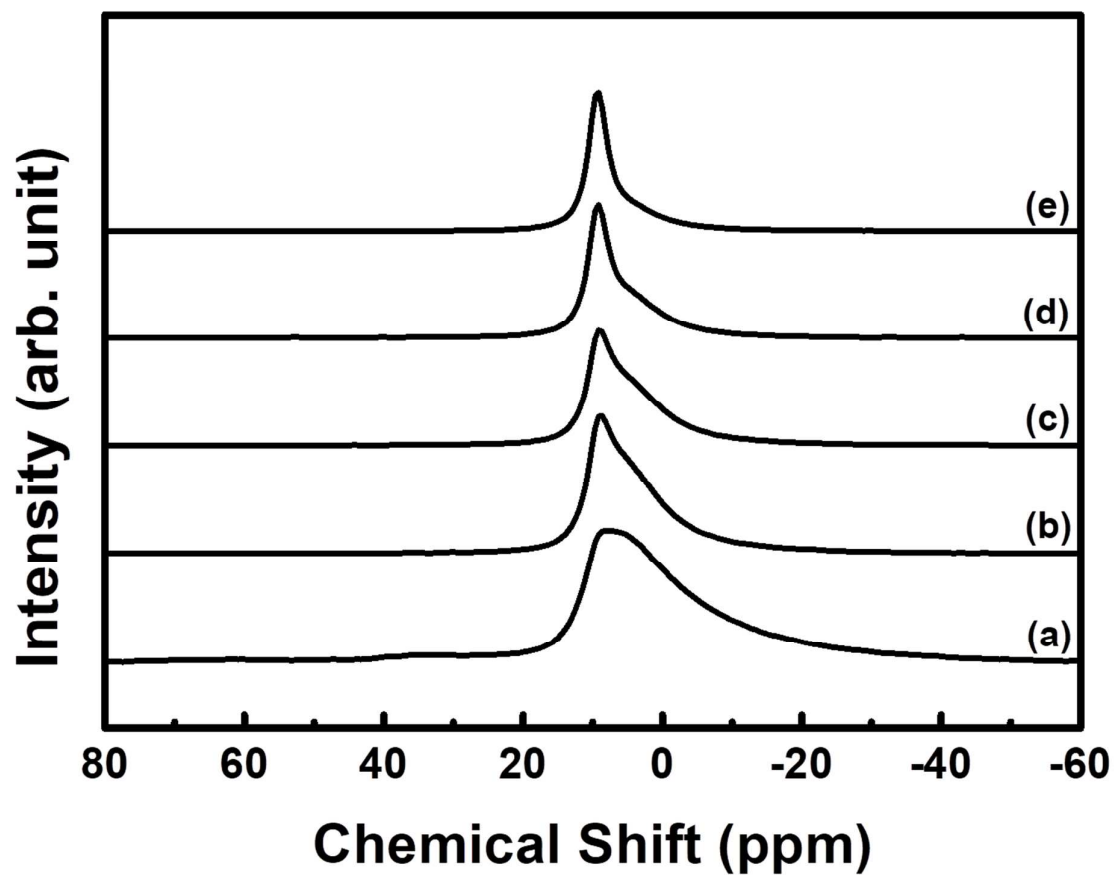
(a) Brucite and hydrothermally obtained precipitates at (b) 1.5 h, (c) 2.3 h, (d) 2.7 h, (e) 3 h, (f) 4 h and (g) 12 h.



**Figure S2.** Calculated aspect ratio ( $\alpha$ ) obtained by crystallite size of (003) and (110) orientation of LDH



**Figure S3.** Scanning electron microscopic images of obtained precipitates of hydrothermally reacted brucite with  $\text{Al}^{3+}_{(\text{aq})}$  after (a) 2.7 h, (b) 3 h and (c) 4 h



**Figure S4.** Solid-state  $^{27}\text{Al}$  MAS NMR spectra for hydrothermally obtained precipitates at (a) 1.5 h, (b) 2.3 h, (c) 2.7 h, (d) 4 h and (e) 12 h. Spectra a, b, c and d were multiplied 4, 3, 2 and 2 times, respectively for appropriate comparison.